

(11) A No. 1067095

(45) ISSUED 791127

(52) CLASS 260-510.8
C.R. CL.

(51) INT. CL. ² C07C 63/26

(19) CA CANADIAN PATENT (12)

(54) PROCESS FOR PRODUCING TEREPHTHALIC ACID

(70) Kimura, Tsuneo; Hashizume, Hiroshi;
Izumisawa, Yoshiaki, Japan

Granted to Mitsubishi Chemical Industries, Limited,
Japan

(21) APPLICATION No. 256,332

(22) FILED 760706

(30) PRIORITY DATE

No. OF CLAIMS 8 - No drawing

PROCESS FOR PRODUCING TEREPHTHALIC ACID

Abstract of Disclosure

There is disclosed a process for producing
terephthalic acid having high purity and suitable for
use in direct esterification with ethylene glycol which
comprises catalytically oxidizing p-xylene in a liquid
phase to obtain a slurry of terephthalic acid while
maintaining the content of 4-carboxybenzaldehyde in the
mother liquor of said slurry less than 800 ppm and
subjecting the slurry to post-oxidation by passing through
molecular oxygen-containing gas therein.

1067095

This invention relates to a process for producing terephthalic acid of high purity and, in more particular, to an improvement of oxidation of p-xylene in a liquid phase to produce terephthalic acid in which p-xylene is catalytically oxidized under specific conditions and a slurry containing terephthalic acid thus obtained is subjected to post-oxidation to obtain high purity terephthalic acid suitable for use in direct esterification with ethylene glycol.

Polyethylene terephthalate which is useful as a raw material for synthetic fiber is obtained by catalytic polymerization of bis-(β -hydroxyethyl)terephthalate and/or its oligomer (hereinafter refers to BHET). BHET is usually produced by either of two ways, that is, by interesterification of p-dialkyl terephthalate and ethylene glycol or direct esterification of terephthalic acid with ethylene glycol. Recently, more interest has been shown in the latter process, but this process requires the use of highly purified terephthalic acid which does not contain more than 500 ppm of 4-carboxybenzaldehyde, the content of 4-carboxybenzaldehyde being a measure of purity of terephthalic acid.

Terephthalic acid produced by the conventional process of oxidizing p-xylene in a liquid phase is contaminated with considerable amounts of undesirable impurities including reaction intermediates such as 4-carboxybenzaldehyde, p-toluic acid and others the structures of which are unknown.

It is essential in such direct esterification to use high grade terephthalic acid which does not contain more than 500 ppm of 4-carboxybenzaldehyde.

It seems possible to obtain such high grade



1067095

terephthalic acid by subjecting a crude terephthalic acid containing a relatively large amount of 4-carboxybenzaldehyde obtained from a conventional process to purification treatment. However, it should be noted that terephthalic acid does not possess a melting point but is decomposed at high temperature and it does not dissolve in almost all of the solvents at normal condition, and therefore terephthalic acid is hardly purified through conventional measure such as distillation and crystallization.

One proposal is found in USP 3584039 in which solid, crude terephthalic acid obtained by oxidizing p-xylene in a liquid phase is dissolved in water at an elevated temperature under superpressure and the aqueous solution is passed together with hydrogen through a fixed bed of palladium supported on active carbon. This process requires to use expensive catalyst with complicated procedures with the result of the increased purification cost and this is not sufficient in commercial point of view.

Alternatively, it seems to reduce the content of 4-carboxybenzaldehyde in terephthalic acid by carrying out the oxidation of p-xylene at a relatively high temperature. However, such high temperature oxidation accompanies the decrease of the yield and contamination of the product. In order to avoid such defects, B.P. 983677 suggests that p-xylene is oxidized in a liquid phase under mild conditions and terephthalic acid slurry thus obtained is contacted with an oxygen-containing gas to effect post-oxidation thereby reducing the content of undesirable impurities. The object of this process is to remove a small amount of impurities, especially coloring materials, remaining in the mother

liquor of terephthalic acid slurry, after the liquid phase oxidation has been completed. Though the purity of terephthalic acid thus treated is considerably improved, the results are still not satisfactory.

5 From our studies, it has been found that the effect of such post-oxidation for reducing the content of impurities depends to considerable extent upon the concentration of 4-carboxybenzaldehyde in the mother liquor of terephthalic acid slurry obtained from liquid phase oxidation of p-xylene.

10 Further, it has also been found that high purity terephthalic acid suitable for direct esterification, that is, the one containing less than 500 ppm of 4-carboxybenzaldehyde, is readily obtained by carrying out the oxidation of p-xylene while maintaining the content of 4-carboxybenzaldehyde in

15 the mother liquor at a predetermined amount by appropriately selecting oxidation conditions and subjecting terephthalic acid slurry thus obtained to post-oxidation.

Accordingly, an object of this invention is to provide a process for producing a high purity terephthalic acid comprising steps of (a) subjecting p-xylene to a one-step oxidation in a lower aliphatic monocarboxylic acid containing 2 to 4 carbon atoms in the presence of an oxidation catalyst comprising heavy metal compound with molecular oxygen-containing gas at a temperature of from 100 to 250°C under a pressure of from atmospheric to 200 atm. to produce a slurry of terephthalic acid and (b) effecting post-oxidation by passing molecular oxygen-containing gas through the slurry obtained from step (a) characterized in that said liquid phase oxidation of step (a) is effected under such condition that the concentration of 4-carboxybenzaldehyde

1067095

in the mother liquor of the slurry is maintained at less than 800 ppm.

By "one-step oxidation" we mean that at least 95%, preferably more than 98% and especially more than 99% by weight of p-xylene is oxidized in a one step to terephthalic acid.

5 This invention will be explained in detail:

In the first step of the process according to this invention, p-xylene is subjected to liquid phase one-step 10 oxidation in a solvent of a lower aliphatic monocarboxylic acid in the presence of an oxidation catalyst containing heavy metal compound.

This liquid phase oxidation is a well established technique, for example, references are made to U.S.P. 15 2833816, 2962361, 3092658, 3115520 and 3119860, and therefore, any conventional process may be employed, provided that reaction conditions, details of which are given hereinafter, are met for the purpose of this invention.

The solvent suitable for the process according to this 20 invention is a lower aliphatic monocarboxylic acid having 2 to 4 carbon atoms such as acetic acid, n-propionic acid and n-butyric acid; and acetic acid is preferred.

The solvent may contain a small amount of water up to 30% by weight and preferably from 10 to 17%. The amount 25 of the solvent to be used is in general from 0.5 to 20 parts by weight per one part of the raw material to be oxidized and preferably 1 to 10 parts.

Examples of the oxidation catalyst which may be used 30 in this process include such as cobalt, manganese, nickel and chromium compounds, especially inorganic salts of cobalt

and manganese capable of being dissolved in the solvent employed and acetate and naphthenate thereof. Such catalyst may be used together with a bromine compound, such as sodium bromide, ammonium bromide, hydrogen bromide and tetrabromoethane. The amount of the catalyst to be used ranges from 0.001 to 10% by weight, in terms of metallic element, of the solvent and that of the bromine compound is from about 50 to about 10000 ppm, in terms of bromine atom, of the solvent.

Examples of the molecular oxygen-containing gas include oxygen and a mixed gas containing oxygen in a proportion more than 5% by volume, such as air. The amount of oxygen to be fed to the reaction system may vary depending upon the type of raw material to be oxidized and ranges in general from 1 to 100 moles per one mole of the raw material and preferably from 3 to 100 moles.

The conditions under which the oxidation is effected are a temperature of from 100 to 250°C and preferably 150 to 250°C, especially 170 to 225°C, and a pressure of from atmospheric to 200 atm., preferably up to 100 atm. and more preferably 6 to 60 atm. However, care should be taken to select the conditions so that the reaction system is maintained in a liquid phase.

According to this invention, the p-xylene is to be oxidized in liquid phase as completely as possible in a one-step. In general, at least 95% by weight of p-xylene is oxidized, preferably more than 98% especially more than 99%.

It is preferable that the liquid phase oxidation be carried out by either continuous process or semi-continuous

process. The oxidation should be controlled in such a manner that the mother liquor of the resulting terephthalic acid slurry contains less than 800 ppm of 4-carboxybenzaldehyde. If the 4-carboxybenzaldehyde content of the mother liquor exceeds the above limitation, more impurities are entrained in terephthalic acid crystals; if such product is subjected to post-oxidation, terephthalic acid of purity is not to be acceptable for use in direct esterification with ethylene glycol. This is made clear by comparing the results of Examples 1 and 2 with those of Comparative Examples 1 and 2.

In Examples 1 and 2, the liquid phase oxidation is carried out in such a manner that the concentration of 4-carboxybenzaldehyde in the mother liquor of the slurry is less than 800 ppm. and then by effecting the post-oxidation there is obtained high purity terephthalic acid containing less than 500 ppm of 4-carboxybenzaldehyde even though the post-oxidation time is shortened as in Example 2.

In contrast, in Comparative Example 1, the liquid phase oxidation time is shortened with the result that the concentration of 4-carboxybenzaldehyde in the mother liquor is 1000 ppm. and the post-oxidation is observed to have little or no effect on improving the purity of the terephthalic acid.

Further, in Comparative Example 2, the oxidation conditions are same as in Comparative Example 1 and post-oxidation time is prolonged and, as a result, although the quantity of impurities in the mother liquor from which precipitated crystals of terephthalic acid have been removed is reduced, the content of impurities in terephthalic

1067095

acid crystals is not changed.

This will be explained as follows.

Most of all terephthalic acid produced in the first
step precipitate in the oxidation vessel and the impurities
5 entrained in crystals are hardly oxidized but only the
impurities retained in the mother liquid.

In view of the above, it will be understood that in
order to obtain high purity terephthalic acid containing
4-carboxybenzaldehyde of less than 500 ppm the concentration
10 of 4-carboxybenzaldehyde of the mother liquor of the tere-
phthalic acid slurry in the oxidation vessel must be held
to less than 800 ppm.

In order to achieve such low concentration of
4-carboxybenzaldehyde in the mother liquor, any of the
15 following measures or a combination thereof is employed in
the oxidation step.

(a) In a continuous process, dwell time of the raw
material in the oxidation vessel is prolonged; for example,
if after one hour dwell time the concentration of 4-carboxy-
20 benzaldehyde is 1600 ppm., then the dwell time is prolonged
to two hours and thereby the concentration is reduced to
less than 800 ppm.

(b) The proportion of the solvent to the raw material
to be oxidized is increased, for example, in a continuous
25 process, it is sufficient to employ a proportion of the
solvent to the raw material of 6 for one hour dwell time.

On the other hand, if the amount of the solvent is
three times the raw material, a dwell time of 2.5 hours is
sufficient.

30 The figures given in (a) and (b) may vary depending

1067095

upon other factors of the reaction conditions.

(c) Water content of the reaction system is lowered.

For example, by means of (1) providing a distillation column to the top of oxidation vessel thereby removing part of the water, (2) discharging part of the distillate from the oxidation vessel into a distillation column from which water is removed while the bottom fraction is recycled to the oxidation vessel, and (3) lowering the water content of the raw material, such as the solvent.

(d) The partial pressure of oxygen in the oxidation vessel is increased, for example, by (1) increasing the amount of oxygen to be fed and maintaining the oxygen concentration at the outlet at 3 to 8 percent by volume and (2) increasing the total pressure in the vessel.

(e) The p-xylene and the molecular oxygen-containing gas are uniformly dispersed throughout the reaction medium, for example, by means of (1) vigorous agitation and (2) separately supplying the p-xylene and the molecular oxygen-containing gas into the oxidation vessel.

Other means, for example, increasing the reaction temperature and using a catalyst having higher activity will also decrease the content of impurities in the mother liquor.

The slurry of terephthalic acid thus obtained is directly subjected to post-oxidation without separating solid terephthalic acid from the slurry.

The object of the post-oxidation is to effect oxidation of impurities, which are oxidation intermediate products and are represented by 4-carboxybenzaldehyde, in the mother liquor, while oxidizing a small amount of unreacted raw material remaining in the slurry simultaneously. Thus it

1067095

is preferable that the slurry to be subjected to the post-
oxidation contains unreacted raw material as small as
possible. If a large amount of unreacted raw material is
present in the slurry, effective post-oxidation cannot be
5 expected. In this connection it is essential according to
this invention that at least 95% by weight of the p-xylene
is oxidized in one-step liquid phase oxidation.

Where the liquid phase oxidation is carried out in a
continuous process, the post-oxidation is effected either
10 in a post-oxidation vessel before transferring into
crystallization vessel or in a crystallization vessel.

Where the liquid phase oxidation is a semi-continuous process,
post-oxidation is effected in the oxidation vessel upon
completion of the oxidation.

15 It is preferable to supply the molecular oxygen-
containing gas in excess of such an amount that reaction
intermediates in the slurry of terephthalic acid are
completely oxidized. However, supplying too much gas
should be avoided, since it causes evaporation of the
20 solvent and water resulting in lowering of the post-
oxidation temperature and then additional heating is
required. If the flow rate of molecular oxygen-containing
gas is appropriately controlled, the post-oxidation of the
slurry of terephthalic acid is effected without heating.

25 The amount of molecular oxygen to be fed to the
post-oxidation step is usually 1/1000 to 1/10 of that fed
to the liquid phase oxidation step since the amount of
impurities to be oxidized is not so large. The molecular
oxygen-containing gas is fed to the liquid phase oxidation
30 step and the post-oxidation step, separately. Usually, the

1067095

oxygen-containing gas is fed in such a rate that the exhaust gas contains from 0.5 to 8% oxygen by volume, preferably 1.5 to 4%. Where the exhaust gas from the liquid phase oxidation step contains a considerable amount of molecular oxygen, such exhaust gas may conveniently be reused for the post-oxidation, if necessary, after mixing with fresh gas. The exhaust from the post-oxidation vessel is discharged to atmosphere after it has been passed through a condenser from which a part or all 5 of the condensate is recycled to the post-oxidation vessel.

The temperature at which the post-oxidation is effected is from 100 to 250°C and preferably from 150°C to that of the liquid phase oxidation. In general, it is convenient to carry out post-oxidation at a temperature 10 between the liquid phase oxidation temperature ($T^{\circ}\text{C}$) and the temperature of ($T-30^{\circ}\text{C}$). Of course, a higher temperature 15 may be employed, but this is uneconomical since further heating is required. The pressure under which post-oxidation is carried out is that the reaction mixture is maintained in a liquid phase.

If necessary, an additional catalyst may be introduced in the post-oxidation step.

The concentration of 4-carboxybenzaldehyde in the mother liquor of the terephthalic acid slurry is measured 20 as follows: from an aliquot of the slurry of terephthalic acid the mother liquor is separated and is subjected to esterification; then the concentration of 4-carboxybenzaldehyde is determined by means of gas chromatography.

This invention will be illustrated further by means 25 of Examples, however, it should be understood that this

1067095

invention is in no way limited by these Examples.

Example 1.

In this Example, continuous liquid phase oxidation was carried out employing as liquid phase oxidation vessel 5 a titanium autoclave equipped with a reflux condenser, a stirrer, a heating means, inlets for gas and solvent containing the raw material and the catalyst and outlets for the slurry of the product and exhaust gas, as post-oxidation vessel a titanium autoclave equipped with a reflux condenser, 10 a stirrer, inlets for the slurry of terephthalic acid and gas and outlets for the slurry and exhaust gas, and a crystallization vessel equipped with a cooling means.

Into the liquid phase oxidation vessel were continuously introduced p-xylene, acetic acid, cobalt 15 acetate tetrahydrate, sodium bromide and water in a proportion by weight of one part, 2.85 parts, 0.004 part, 0.008 part, 0.0036 part and 0.15 part, respectively, while air was passed through the mixture and agitation was 500 r.p.m. to effect the oxidation. The oxidation reaction 20 was controlled in such a manner that the dwell time of the p-xylene in the vessel was 175 minutes and the exhaust gas contained 4% by volume of oxygen while the reaction temperature and pressure were maintained at 210°C and 25 kg/cm², respectively.

The resulting slurry of terephthalic acid was fed 25 into the post-oxidation vessel in which the post-oxidation was effected at a temperature of 190°C under a pressure of 17.5 kg/cm² for a dwell time of 85 minutes while there was introduced a mixture of air and the exhaust gas from the 30 liquid phase oxidation step (the oxygen concentration thereof

1067095

being 10% by volume) at such a rate that the exhaust gas contained 4% by volume of oxygen.

5 The slurry of terephthalic acid was transferred to the crystallization vessel maintained at 100°C and the crystals precipitated were separated.

The terephthalic acid thus recovered was suspended in 4 times by weight of acetic acid, stirred at 80°C for 20 minutes, separated and dried.

10 Properties of the resulting terephthalic acid, the mother liquor recovered from the oxidation vessel and the mother liquor recovered from the crystallization vessel are given in Table 1, Table 2 and Table 3, respectively.

Example 2.

15 The procedures similar to those of Example 1 were repeated excepting that in the liquid phase oxidation the dwell time was 80 minutes, and in the post-oxidation the dwell time was 45 minutes at a temperature of 190°C and under pressure of 16 kg/cm².

20 The results are given in Tables 1 to 3.

Example 3.

25 The procedures of Example 1 were followed but in the liquid phase oxidation 6 parts of acetic acid and 109 minutes dwell time were employed and in the post-oxidation 53 minutes dwell time, 195°C of temperature and 17.5 kg/cm² of pressure were employed.

The results are given in Tables 1 to 3.

Example 4.

30 Terephthalic acid was produced following the procedures similar to those of Example 2 but the liquid phase oxidation was effected at a temperature of 220°C

1067095

under a pressure of 29 kg/cm².

The results are given in Tables 1 to 3.

Comparative Example 1.

The procedures of Example 1 were followed but the
5 dwell time in the liquid phase oxidation vessel was
prolonged to 57 minutes.

The results are given in Tables 1 to 3.

Comparative Example 2.

The procedures of Comparative Example 1 were
10 repeated but the dwell time in the post-oxidation was
prolonged to 180 minutes.

The results are given in Tables 1 to 3.

Comparative Example 3.

Terephthalic acid was produced by the liquid phase
15 oxidation according to Example 1 but no post-oxidation was
effected.

The properties of terephthalic acid thus obtained
are given in Table 1.

Comparative Examples 4 to 6.

The procedures similar to those of Example 1 were
repeated excepting that the amount of water to be fed to
the oxidation vessel was increased from 0.15 part to 0.45
part (Comparative Example 4), the rate of agitation was
changed from 500 r.p.m. to 200 r.p.m. (Comparative
25 Example 5) and the concentration of oxygen in the exhaust
gas was changed from 4% by volume to 1% (Comparative
Example 6).

The results are given in Tables 1 to 3.

Reference Example.

30 Polyethylene terephthalates were produced by

esterification of terephthalic acids obtained by Examples 1 to 4 and Comparative Examples 1 to 6 with ethylene glycol. Terephthalic acid (4.98 kg), ethylene glycol (3.72 kg), antimony trioxide (0.875 g), cobalt acetate tetrahydrate (0.747 g) and trimethyl phosphate (1.344 g) were charged to an autoclave which was then purged with argon, and esterification was conducted at a temperature of 235°C under a pressure of 2.5 kg for 4 hours with agitation being a power of 0.4 KW while water formed was continuously removed; then the polymerization conditions of a temperature at 280°C and a pressure less than 1 torr. were set over a period of 45 minutes and these conditions were maintained until the power for agitation increased by 0.13 KW.

The polyester thus produced was discharged into water and cut to form chips 2 mm in diameter and 3 mm in length.

The results are given in Table 4.

The saturation (Hunter b-value) of products of polyethylene terephthalate was measured by Color Machine Model DC-5D available from Tokyo Denshoku Kabushiki Kaisha, Tokyo, Japan. "b-Value" is a measure indicating that the larger the figure the product is more yellowish.

1067095

Table 1.
Properties of terephthalic acid

| No. | Example | | | | Comparative Example | | | | | |
|---|---------|-------|-------|-------|---------------------|-------|-------|-------|-------|-------|
| | 1 | 2 | 3 | 4 | 1 | 2 | 3 | 4 | 5 | 6 |
| Purity (% by weight) | 99.96 | 99.94 | 99.96 | 99.96 | 99.90 | 99.90 | 99.88 | 99.90 | 99.90 | 99.90 |
| 4-CBA (1) (% by weight) | 0.030 | 0.048 | 0.025 | 0.031 | 0.075 | 0.075 | 0.090 | 0.070 | 0.060 | 0.075 |
| Ash (% by weight) | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 |
| Transmission (2) (% at 340 m μ) | 93.0 | 91.0 | 93.5 | 92.0 | 87.0 | 87.0 | 85.0 | 90.0 | 80.0 | 85.0 |
| Transmission (3) (% at 400 m μ) | 98.0 | 98.0 | 98.0 | 98.0 | 97.5 | 97.5 | 97.0 | 97.8 | 96.0 | 97.1 |

Note:

- (1): 4-Carboxybenzaldehyde.
- (2) and (3): measured by a quartz cell of a ray length 1 cm using a solution of 15 parts of terephthalic acid in 100 parts of 2N-potassium hydroxide

1067095

Table 2
Properties of the mother liquor of the slurry
 in the liquid phase oxidation vessel

| No. | Example | | | | | Comparative Example | | |
|--------------------|---------|------|------|------|---------|---------------------|------|------|
| | 1 | 2 | 3 | 4 | 1 and 2 | 4 | 5 | 6 |
| 4-CBA (1) (ppm) | 300 | 750 | 250 | 500 | 1000 | 950 | 900 | 1050 |
| PTA (2) (ppm) | 1600 | 3500 | 1300 | 2200 | 4800 | 3800 | 3600 | 4500 |

Note: (1) 4-Carboxybenzaldehyde
 (2) Para toluidic acid

Table 3

Properties of the mother liquor of the slurry
 after the crystallization

| No. | Example | | | | | Comparative Example | | |
|-------------|---------|-----|-----|-----|-----|---------------------|-----|-----|
| | 1 | 2 | 3 | 4 | 1 | 2 | 4 | 5 |
| 4-CBA (ppm) | 70 | 130 | 70 | 80 | 140 | 60 | 140 | 130 |
| PTA (ppm) | 150 | 210 | 150 | 150 | 250 | 140 | 220 | 220 |

1067095

Table 4

| | Terephthalic acid produced by | Hunter b-value |
|---------------------|---|--|
| Example | 1 2 3 4 | 3.5 3.8 3.4 3.6 |
| Comparative Example | 1 2 3 4 5 6 | 4.7 4.7 5.0 4.2 5.8 5.0 |
| | Purified terephthalic acid ⁽¹⁾ | 3.0 |

Note: (1) A crude terephthalic acid was dissolved in water at high temperature under superpheric pressure and treated with hydrogen in the presence of a palladium catalyst.

1067095

The embodiments of the invention in which an exclusive property or privilege are claimed are defined as follows.

1. In a process for producing high purity terephthalic acid comprising steps of:

- (a) subjecting to a one-step oxidation p-xylene in a lower aliphatic monocarboxylic acid as a solvent with a molecular oxygen-containing gas in the presence of an oxidation catalyst containing heavy metal compound to obtain a slurry of terephthalic acid, and
- (b) subjecting the slurry of terephthalic acid to post-oxidation by passing molecular oxygen-containing gas through the slurry,

the improvement in which said liquid phase oxidation of step (a) is effected while the concentration of 4-carboxybenzaldehyde of the mother liquor in the slurry is maintained at less than 800 ppm.

2. A process for producing terephthalic acid according to claim 1, wherein said post-oxidation is effected at a temperature that of the liquid phase oxidation or lower.

3. A process for producing terephthalic acid according to claim 1, wherein said solvent is acetic acid.

4. A process for producing terephthalic acid according to claim 1, wherein said oxidation catalyst is cobalt compound and/or manganese compound.

5. A process for producing terephthalic acid according to claim 1, wherein said oxidation catalyst containing heavy metal compound is used together with a promoter of bromine compound.

6. A process for producing terephthalic acid according to claim 1, wherein said liquid phase oxidation of

1067095

step (a) and said post-oxidation of step (b) are effected continuously.

7. A process for producing terephthalic acid according to claim 1, wherein said molecular oxygen-containing gas in step (b) is the exhaust gas from step (a) alone or a mixture of fresh air therewith.

8. A process for producing terephthalic acid according to claim 1, wherein any of following measures or a combination thereof is applied to said liquid phase oxidation of step (a) whereby a content of 4-carboxybenzaldehyde less than 800 ppm in the mother liquor of the terephthalic acid slurry is achieved:

- (1) in a continuous process, the dwell time of the raw material in the oxidation vessel is prolonged,
- (2) the proportion of the solvent to the raw material to be oxidized is increased,
- (3) the water content of the reaction system is lowered,
- (4) the partial pressure of oxygen in the oxidation vessel is increased, and
- (5) the p-xylene and the molecular oxygen-containing gas are uniformly dispersed throughout the reaction medium.

Patent Agents:

George H. Riches and Associates
2 Bloor Street East, Suite 2900
Toronto, Ontario M4W 3J5 Canada